775 **Supplementary Materials**

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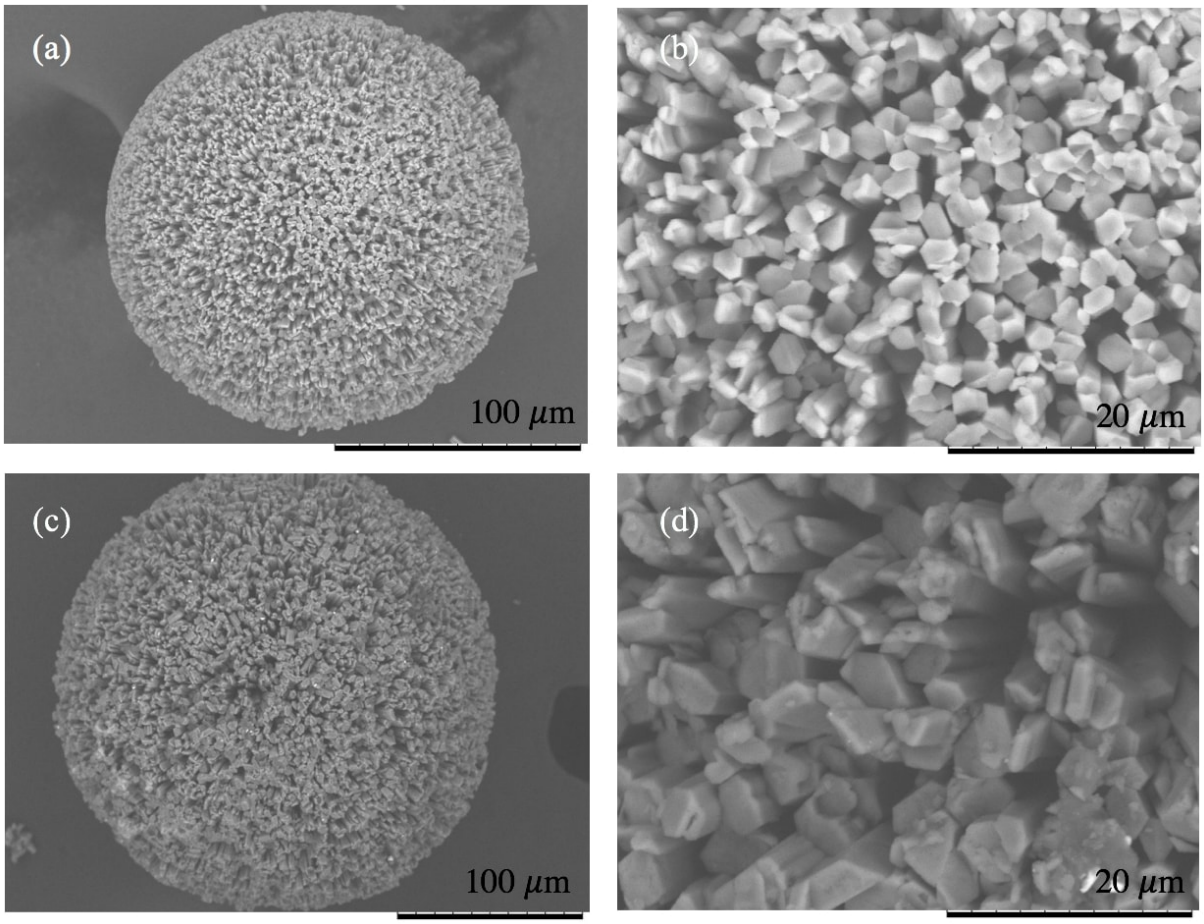

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778 Fig. S1 SEM image of ^{13}C-labeled U-tube aragonite samples (a, b) and the samples post-
779 dissolution (c, d) (after 17 days in seawater of Ω=0.64, ~6% of the original samples were
780 dissolved). Samples are spherical aggregation of needles with hexagonal cross-sections.

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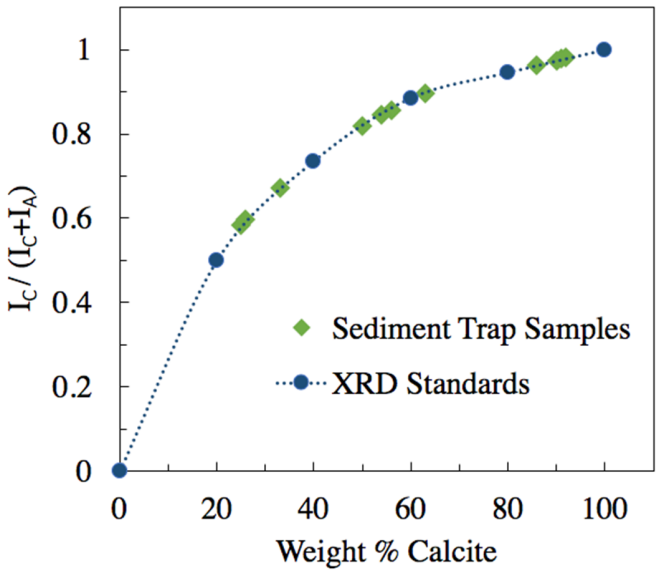

Fig. S2 Relative peak intensity of calcite (104) and aragonite (111) XRD peaks vs. calcite percentage. I_C is the peak intensity of calcite (104) peak, I_A is the peak intensity of aragonite (111) peak. Blue circles are standards with different ratios of calcite and aragonite. Green diamonds are sediment trap samples. A set of 6 standards (blue circles) was prepared by mixing different weight % of calcite and aragonite. A standard curve (blue dotted line) of the relative peak intensity vs. weight % calcite was generated. Calcite percentages in the sediment trap samples (green diamonds) were estimated by fitting the data points in the standard curve.

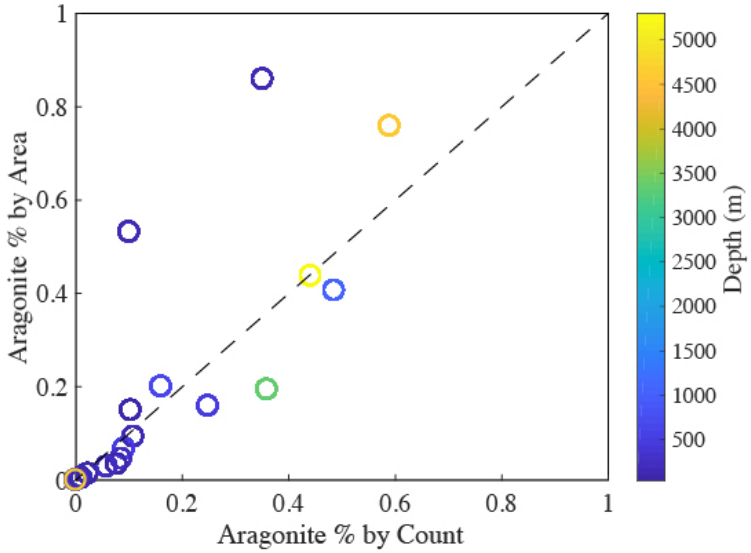

Fig. S3 Analysis of aragonite percentages in suspended carbonate. Aragonite percentages calculated based on counts and area of calcite/aragonite on the filters generally agree, except for several filters that captured large aragonite or calcite particles. The data points above the 1:1 line in this plot indicate filters that include large aragonite fragments, whereas the data points below the 1:1 line indicate filters that include large calcite fragments. The inclusion of single large particle of either mineral phase is random and not related to depth.

Table S1 A comparison of Ω values determined by USC (lab method in this paper) and USF (field method in this paper) by measuring the same seawater from SPOT (San Pedro Ocean Time-Series)

Depth (m)	USC			USF		
	DIC (μmol/kg)	Alkalinity (μmol/kg)	Ω	pH_{Total}	Alkalinity (μmol/kg)	Ω
250	2250.6	2271.6	1.12	7.4097	2273.6	1.17
500	2308.9	2303.6	0.88	7.3306	2303.0	0.91
750	2350.1	2342.1	0.82	7.3117	2344.6	0.83

* Alkalinity was measured at 21°C; pH was measured at 25°C

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807 Table S2 Aragonite dissolution experimental conditions and rates

In situ dissolution in the North Pacific					
Station #	Depth (m)	Temperature (℃)	$\Omega_{aragonite}$	Dissolution rate ($\times 10^{-3}$ g g^{-1} day^{-1})	Rate error ($\times 10^{-3}$ g g^{-1} day^{-1})
Station 2	700	5.433	0.557	3.86	0.35
	710	5.329	0.540	4.02	0.35
	2020	2.092	0.571	5.26	1.47
Station 3	500	6.138	0.804	1.60	0.08
	575	5.190	0.671	2.07	0.11
	600	4.983	0.638	1.79	0.14
	650	4.694	0.552	7.39	0.18
	700	4.421	0.452	4.53	0.22
	730	4.281	0.488	6.52	0.08
	740	4.232	0.486	5.24	0.27
Station 4	350	6.913	0.965	0.99	0.15
	400	6.114	0.799	1.17	0.26
	425	5.687	0.767	1.73	0.12
	450	5.387	0.721	2.48	0.12
	500	4.995	0.642	3.55	0.21
Station 5	100	4.185	1.199	0.84	0.32
	110	3.981	1.033	0.66	0.26
	115	4.079	0.944	0.72	0.23

	120	4.049	0.868	0.94	0.29
	50	6.464	1.464	0.46	0.28

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Laboratory dissolution

Expt. #	Temperature (°C)	$\Omega_{aragonite}$	$\Omega_{aragonite}$ error	Dissolution rate ($\times 10^{-3}$ g g^{-1} day^{-1})	Rate error ($\times 10^{-3}$ g g^{-1} day^{-1})
A3	21	0.867	0.029	1.78	0.07
A4		1.009	0.016	1.19	0.08
A5		0.828	0.019	3.01	0.08
A6		0.971	0.022	0.70	0.08
A7		0.878	0.007	1.71	0.05
A8		0.652	0.017	8.30	0.44
A9		0.558	0.010	13.83	0.53
A10		0.551	0.013	13.84	0.02
A11		0.515	0.006	16.68	0.07
A14		0.636	0.016	6.74	0.15
A17		0.710	0.017	4.82	0.15
A15	5	0.685	0.012	4.76	0.50
A18		0.672	0.023	2.48	0.07
A19		0.699	0.016	1.80	0.06
A20		0.539	0.012	10.07	0.59
A21		0.650	0.008	1.66	0.15
A22		0.818	0.020	1.78	0.07

A23		0.543	0.006	4.98	0.09
A24		0.642	0.010	2.61	0.09
A26		0.587	0.016	4.52	0.08
A27		0.743	0.017	3.14	0.09

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811 Table S3 Carbonate percentages by count and by area on *in situ* pump filters

Filter ID	Depth (m)	Latitude	Calcite/$CaCO_3$		Note
			By count	By area	
S1-D1-1	4575	22°45'N	91%	63%	Large aragonite fragment
S1-D1-3	4575		41%	24%	
S1-D2-1	3375		64%	81%	
S1-D3-1	150		89%	91%	
S2-D1-1	1100	27°45'N	51%	59%	
S2-D2-1	620		84%	80%	
S2-D3-1	450		91%	93%	
S2-D4-1	150		100%	100%	
S2-D4-2	150		90%	47%	Large aragonite fragment
S2-D6-1	5300		56%	56%	
S2-D7-1	40		99%	100%	
S3-D1-1	1000	35°16'N	N.A.	N.A.	No $CaCO_3$ detected
S3-D2-1	400		100%	100%	
S3-D3-1	250		91%	95%	
S3-D4-1	150		99%	100%	
S3-D5-1	5100		100%	100%	
S3-D6-1	75		98%	100%	
S3-D6-2	75		99%	99%	
S4-D2-1	450	41°45'N	100%	100%	
S4-D3-1	150		65%	14%	Large aragonite fragment

S4-D4-1	4700		99%	99%	
S4-D5-1	35	49°50'N	92%	97%	
S5-D1-1	500		75%	84%	
S5-D2-1	200		94%	97%	
S5-D3-1	100		90%	85%	
S5-D5-1	4500		N.A.	N.A.	No $CaCO_3$ detected

812 * Filter ID were named after: Station # - Depth # - Triplicate #

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Model of aragonite dissolution fluxes in the North Pacific water column

(1) Thermodynamically-driven (abiotic) aragonite dissolution

We construct a box model of the water column to diagnose the sinking and dissolution of $CaCO_3$ particles in the upper 2000 m. First, to estimate the dissolution flux due to abiotic aragonite dissolution, sinking aragonite particles are considered to be exposed to dissolution when the water column first reaches $\Omega < 1$, and dissolution rates are determined as a function of Ω according to the relationship shown in Fig. 2. The whole water column is separated into multiple boxes (box height from 20 to 800 m depending on the depth of CTD deployment). The amount of aragonite dissolution in each box depends on the aragonite flux that reaches the box, the average saturation state of the box, and the aragonite particles' residence period in the box. The sinking flux in the ith box $Flux_{Zi}$ (mmol m^{-2} day^{-1}) and *in situ* dissolution rate in the ith box *In Situ Rate*$_{Zi}$ (μmol kg^{-1} yr^{-1}) are calculated based on the following equations:

$$Flux_{Zi} = Flux_{Zi-1} - Flux_{Zi-1} * Dissolution\ Rate * time$$

$$= Flux_{Zi-1} - Flux_{Zi-1} * [0.013(1-\Omega_i)^{1.37}] * \frac{(Z_i - Z_{i-1})}{w} \quad \text{Eq. 1}$$

$$In\ Situ\ Rate_{Zi} = \frac{Flux_{Zi} * S * [0.013(1-\Omega_i)^{1.37}] * \frac{(Z_i - Z_{i-1})}{w}}{S * (Z_i - Z_{i-1}) * \rho}$$

$$= \frac{Flux_{Zi} * [0.013(1-\Omega_i)^{1.37}]}{\rho * w} \quad \text{Eq. 2}$$

where Z_i (m) is the depth of the ith box, $Flux_{Zi}$ (mmol m^{-2} day^{-1}) is the sinking flux of aragonite at depth Z_i, Ω_i is the saturation state of water column at Z_i, w (m day^{-1}) is the sinking rate of aragonite flux, $In\ Situ\ Rate_{Zi}$ (μmol kg^{-1} yr^{-1}) is the *in situ* dissolution rate of aragonite at depth Z_i, S (m^2) is the area of the water-column cross section, and ρ (1029 kg m^{-3}) is the density of seawater. In Eq. 1 and 2, aragonite dissolution rate is calculated as established by our dissolution experiments: Rate (g g^{-1} day^{-1}) = 0.013*(1- Ω)$^{1.37}$.

To estimate the average aragonite dissolution fluxes in the North Pacific, water column saturation is taken from measurements at Station 3 (35°N, 151°W); and an aragonite sinking flux of 0.1 mmol m^{-2} day^{-1} is assumed at the saturation horizon (430 m). Values adopted in the model are chosen to be generally representative of the N. Pacific transect.

(b) Respiration-driven or metazoan/zooplankton consumption-driven PIC dissolution

For the shallow depth dissolution indicated by PIC losses in the sediment trap samples, a model is developed to simulate the process in which sinking fluxes at 100 m are taken from measurements using sediment traps, and a constant dissolution rate of 40% every 100 m is assumed based on the calculated dissolution rate between 100 and 200 m (Fig. 7b). The sinking flux (mmol m^{-2} day^{-1}) and the *in situ* dissolution rate (μmol kg^{-1} yr^{-1}) are calculated based on the following equations:

$$Flux_{Zi} = Flux_{Zi-1} - Flux_{Zi-1} * \frac{40\%}{100m} * (Z_i - Z_{i-1}) \quad \text{Eq. 3}$$

856 $$In\ Situ\ Rate_{Zi} = \frac{Flux_{Zi} * S * \frac{40\%}{100m} * (Z_i - Z_{i-1})}{S * (Z_i - Z_{i-1}) * \rho} = \frac{Flux_{Zi} * 40\%}{100m * \rho}$$

857 Eq. 4

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859 PIC, calcite and aragonite sinking fluxes at 100 m ($Flux_{100m}$) are 1.2 mmol m^{-2} day^{-1}, 0.8 mmol

860 m^{-2} day^{-1}, and 0.4 mmol m^{-2} day^{-1}, respectively.

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